

X-RAY POLE FIGURE ANALYSIS ON FIBER TEXTURED EPITAXIAL NIOBIUM FILMS FOR SRF CAVITIES*

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Abstract

Single and large grain Nb films are of interest due to their potential for reduced cost to replace bulk Nb SRF cavities. The structural properties and SRF performance of Nb films obtained by coaxial energetic deposition (CEDTM) in a cathodic arc vacuum process are compared and discussed. The CEDTM is a hybrid technique with both energetic ion deposition and implantation phase based on cathodic arc plasma sources, which are copious generators of condensable energetic (20-200 eV), multiply charged ions from metal or alloy cathodes. X-ray diffraction (XRD) pole figures are used to investigate the grain orientations on Nb films grown at different substrate temperatures. The pole figures indicate good structural and electrical properties. The ratio of residual resistivity (RRR) dependence on a-plane sapphire substrate temperature shows RRR of ~129 and $T_c=9.2K$ for 400 °C substrate, dropping to only ~4 for a room temperature substrate. This RRR figure of merit suggests good SRF performance in such Nb thin-films.

INTRODUCTION

The growth behavior of niobium films on sapphire substrates has been studied in detail and the epitaxial layers are well understood to exhibit almost strain free and large structural coherence length [1]. The crystallographic system is quite interesting from a physics viewpoint, as these thin-films differ significantly from bulk Nb in terms of their transport, thermo-dynamical [2], and structural [3-5] properties.

In this paper, we report on x-ray texture studies that explore the relationship between the crystal structure and electrical conductivity under different growth conditions. Single crystalline niobium films on a-plane sapphire prepared by a coaxial energetic depositionTM (CEDTM) process [6] at various substrate temperature have been attracting considerable attention as low cost and stable materials for thin film SRF cavities [7-8]. The microstructures of the crystalline phase are correlated with crystallographic preferential orientation. The (110) preferentially oriented Nb films contain fiber textured

microstructures, which have been revealed by pole figure x-ray diffraction (XRD) measurements in

conjunction with the crystal structure studied by the electron backscatter diffraction (EBSD) technique. The difference in the crystal structure is related to the device performance. Specifically, concerning the grain orientation at 400 °C, the (110) preferentially oriented Nb films have been believed to be advantageous mainly from the aspect of the untwining dislocation and reducing stress. The contribution of different types of defects to the residual resistivity of Nb films can be determined by impurities, dislocations, and grain boundaries.

Characterization tools used were the ratio of residual resistivity (RRR) and superconducting transition temperature (T_c) to investigate superconducting radio frequency (SRF) performance. Crystalline and grain orientations were measured by electron beam scattering diffraction (EBSD) and x-ray diffraction (XRD). Pole figure measurements at a diffracted position were performed to determine the average disorientation angles of subgrains. Texture formation in thin films directly influences the physical characteristics of materials such as magnetic, mechanical and electrical properties.

X-RAY DIFFRACTION ANALYSIS

X-ray diffraction is one of the most powerful and widely used techniques for accurate characterization of the lattice parameters, mismatch, and thickness of epitaxial materials. Bragg's law may be expressed in vector notation. Let \vec{K}_i , \vec{K}_d be unit vectors along the directions of the incident and diffracted beams, then the scattering vector, $\vec{S} = \vec{K}_d - \vec{K}_i$, is parallel to \vec{d}_{hkl}^* , the reciprocal lattice vector of the crystal reflecting planes. Comparing the moduli of these vectors $|\vec{S}| = 2 \sin \theta$

and $|\vec{d}_{hkl}^*| = 1/d_{hkl}$, it is seen from Bragg's law that their ratio is simply the wavelength λ . The scattering vector of the conventional 2θ - ω scans in the reciprocal space is shown in Fig 1 (a). The scans move along the sample surface normal direction, which includes the information of the interplanar space in the crystal. If the Nb films are grown as a single (or poly) crystalline structure, all possible diffraction peaks in the 'out-of-plane' orientation can be collected by the 2θ - ω scans. The pole figure is one of the most widely used methods for identification of crystal orientation in the film. The measurement can

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collect the intensity distribution in the reciprocal space over the surface of a hemisphere that has a radius of scattering vector \vec{S} shown in Fig 1 (b).

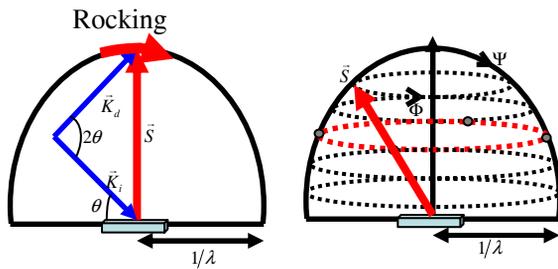


Figure 1: (a) schematic of scan vector of 2θ - ω scan and x-ray rocking curve in reciprocal space (b) schematic drawing of pole figure measurement in reciprocal space.

Information on the inclination angle of the equivalent diffraction or information on the angle from the oriented crystallographic plane can be obtained by this method. Whereas a standard 2θ - ω XRD scan measures only grains with Bragg planes *parallel* to the film plane, the texture analysis shows if the sample is tilted against the incidence plane of the X-ray beam and rotated around the sample normal to get all grains in a diffracting position. The resulting intensities are represented by two-dimensional pole figures and interpreted as distribution of crystalline orientations.

X-RAY PATTERN OF NB FILMS

Nb films deposited on a-plane sapphire substrate at various substrate temperatures are crystalline as shown by the XRD pattern from 30 to 100° (2θ - ω scan) in Fig 2. The Bragg-Brentano scans include information on the d-spacing, where d is the interplanar spacing of successive atomic planes in the crystal that lie along the surface normal direction. When only the diffraction peak of the (110) plane and of the next diffraction order of (220) were detected, these deposited layers show a preferred orientation of the Nb {110} plane parallel to the substrate. XRD measurements on these Nb samples show sharp and narrow Bragg diffracted peaks, with the diffraction vector perpendicular to the substrate surface. The primary peaks visible for all Nb samples show the {110} peak corresponding to the bcc phase. Since the peak width is inversely proportional to the XRD crystallite size, the increase in peak width indicates a reduction in the mean crystallite size, based on Debye-Scherrer broadening. All samples are clearly observed to show their peaks near the 38.6° and 82.6° angles, which correspond to (110) and (220) phases, respectively. The peak position for the 400°C substrate temperature was $2\theta=38.4^\circ$ with 0.255° full-width at half maximums (FWHM), which indicates a well crystallized material (near bulk-like) and a strain of 0.3% or less with respect to bulk Nb.

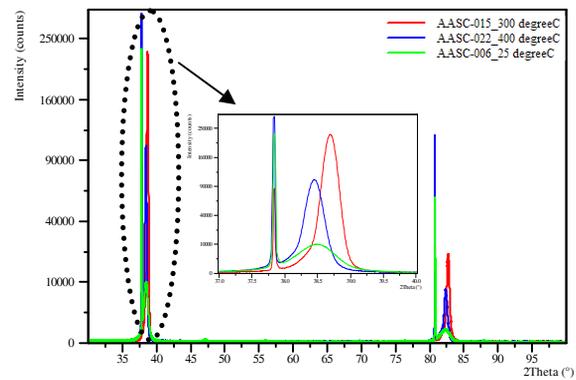


Figure 2: XRD patterns of Nb films on a-plane sapphire, coated at 400°C (red), 300°C (blue), and room temperature (green).

IN-PLANE TEXTURE MEASUREMENT

The variation in degree of crystal orientation was measured by x-ray pole figure analysis. By measuring the intensity of the (110) Bragg diffraction peak ($2\theta=38.24$ for Cu $K\alpha$ radiation) over almost a full hemisphere, from the substrate normal to an angle of 85° from normal, the distribution of poles is obtained and displayed on a stereographic projection. A film with random orientation would give a uniform distribution of scattering intensity. A single crystal film displays a spot pattern, and a film with restricted fiber texture displays a ring pattern of modulated intensity, tending toward a spot pattern with increasing degree of orientation. A film with fiber texture, but random azimuthal distribution of grains, displays a ring pattern of uniform intensity. Therefore, the pole figure data may be displayed as the diffracted intensity as a function of the angle of rotation and tilt of the sample. Texture measurement of three of Nb films grown on a-plane sapphire substrates at different substrate temperatures is shown in Fig 3. The pole figure of Nb film deposited on a-plane sapphire substrate at 400°C temperatures is shown in Fig 3 (a). The central spot indicates the grains oriented with (110) planes parallel to the substrate, while four spots at 60° of tilt correspond to diffraction intensities from {110} in-plane texture. The azimuthal angles of these four intensity maxima are 42.5° , 153° , 223° , and 333.5° , respectively, and are separated by angles of 70.5° and 109.5° . As shown in Fig 3 (b), the degree of {110} in-plane texture shows the diffraction intensity as a function of rotation (Φ -scan) and 60° tilt sample, that indicates a strong (110) fiber texture perpendicular to the substrate. The detector was set for the (110) reflection from the Cu $K\alpha$ line and the sample scanned about the rotation axis. Four peaks were detected separated by 109.5° and 70.5° . The size of the specimen was such that these could not arise from bulk scattering from the edge of the sapphire and represent Nb surface diffraction peaks. In Fig. 3 (c), the crystal orientation mapping of Nb film using SEM/EBSD shows the uniform green color (see inset triangle) and a single spot in the Kikuchi pattern that indicates a single {110}

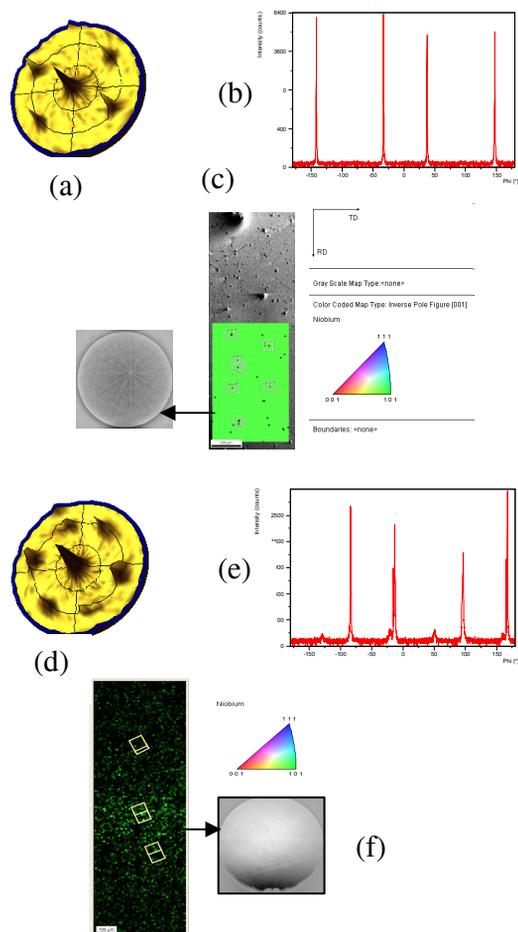


Figure 3: X-ray pole figure and EBSD images of (110) Nb films on a-plane sapphire substrate (a-c) at 400°C substrate temperature, (d-f) at 20°C substrate temperature.

orientation plane. At room temperature in Fig 3 (d), the diffraction intensity at the center spot indicates the grains are strongly oriented with (110) planes parallel to the substrate, but in-plane texture shows six spots at 60° tilt. The figure shows two groups of (110) Nb poles: one group located 57.5°, 169°, 237.5°, and 343°, with peaks separated by angles of 71.5±2° and 108.5±0.5°, respectively; the other group has maximum intensity at 103°, 169°, 283°, and 343°, with these angles separated by 63±3° and 117±3°. This means that (110) epilayers films are well adhered to the a-plane substrate, but there are two kinds of twins that are rotated by 45° about the film normal. The maximum intensity of these two groups in Fig 3 (e) means that the film has a small amount of twins, because the relative intensities are quite different. This is called screw dislocation; the crystal is not made up of parallel atomic planes one above the other; rather it is a single atomic plane in the form of a spiral ramp. The slip vector of the dislocation is parallel to the dislocation line for a screw dislocation. In Fig. 3(f), the EBSD image and

the Kikuchi pattern show poor quality because of local geometrical roughness.

DISCUSSION

The value of RRR of a superconducting thin-film is determined by the complex interplay between impurities, dislocations, and grain boundaries. To measure RRR of Nb film on sapphire substrate, using a four-point-probe technique, one measures the electrical resistivity by a ratio of the differential voltage which is dropped at room temperature, 300K, and just above the transition at 10K, respectively. The measurement of the RRR values for 400 °C, 300 °C, and room temperature gives values of 129, 50, and 4 respectively. The influence of dislocation on the value of RRR is related not only to their crystal orientation but also to their grain distribution. RRR is observed to drop dramatically at lower substrate temperature. X-ray pole figures suggest screw (or twin) dislocations are formed at lower substrate temperatures, which in turn might be responsible for the observed lower RRR values.

The motion of screw dislocation can be described by molecular dynamics (MD) as the magnitude of stress and temperature in bcc phase [9]. The dislocations move smoothly through the formation and migration of atomic sized kinks under low stress. As stress is further increased, the motion of dislocation becomes rough and jerky in its motion vector. Under still higher stress, the line roughness is increased continually until at some point is initiated a thin plate of sheared crystal; *i.e.* a twin. The twin starts where the dislocation motion just left off, then grows rapidly along the same direction as that of the dislocation motion. Although experimental results suggest an angle of 45°, we can calculate the twin disorientation $\theta=39^\circ$ by using $\theta \approx 4 \tan^{-1} \left[\frac{a-b}{a+b} \right]$, where a and b are the lattice parameters in bulk Nb material.

CONCLUSIONS

We have investigated the growth, structure, and the electrical conductivity of Nb films on a-plane sapphire using the CEDTM process. XRD and EBSD experiments show that these samples are single crystal in structure but strongly textured. Pole figures from XRD measurements contain not only information about the crystal orientation and the distribution of the planes along the substrate surface but also the grain orientations and the dislocations. The change in resistivity due to the twin dislocation is measured by the uniformity of grain distribution that can lead to the decrease of the value of resistivity due to such dislocations.

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